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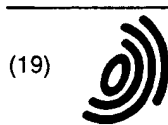
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(54) **Resin-free succinimides**

Harzfreie Succinimide

Succinimides sans résines

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## Description

The present invention relates to novel pure hydrocarbyl substituted succinylating agents and substantially pure derivatives thereof, to a method for the preparation of the hydrocarbyl substituted succinylating agents and the derivatives, and to the use thereof as additives for fuel and lubricants.

Additives are an important means of maintaining engine performance either by cleaning and/or lubricating moving parts of the engine. It is well known in the art to prepare hydrocarbyl substituted succinylating agents (hereafter "HSSA"), and specifically poly(iso)butenyl succinic anhydrides (hereafter "PIBSAs") by the reaction of a poly(iso)butene with maleic anhydride and to convert the PIBSAs to the corresponding succinimides. These succinimides are used as fuel additives in view of their ability to clean and/or maintain in a clean condition carburettors, manifolds, inlet valve ports and the like in an internal combustion engine. However, the PIBSAs and the corresponding succinimides produced by conventional routes, eg by reaction with maleic anhydride and the subsequent reaction of the PIBSA with a polyamine, normally contain undesirable resins which not only show a significant discrepancy between the real and apparent succinylation in the maleinised product (due to the presence of resinified and/or unreacted maleic anhydride) but also adversely affect the detergency properties of such additives. Moreover, the method of manufacturing these additives by the intermediate chlorination route is also undesirable because the presence of any chlorine in the succinimide product may give rise to undesirable products such as dioxin under the conditions prevalent in an internal combustion engine. In addition, the presence of chlorine is, in any case, environmentally undesirable should such products or the packaging containing such products be discharged into the environment accidentally or as waste.

For instance, GB-A-1483729 which relates to a process for producing an ester of PIBSA with a polyvalent alcohol, describes preparation of PIBSA by the thermal maleinisation of polyisobutene with maleic anhydride followed by washing of the resultant product with a mixture of water and a monoalcohol. However, the washed product still has an acid number of 0.85 milli-equivalent residual acid per gram which signifies the presence of acidic impurities in the washed product. This reference makes no mention of the chlorine content of the washed PIBSA. Similarly, FR-A-2321508 (equivalent to GB-A-1543039) describes a process for producing PIBSA by initial thermal maleinisation of polyisobutene with maleic anhydride followed by chlorination of the reaction mixture at elevated temperature. The resultant product is then diluted with white spirit and then rendered free of tarry products by washing with a mixture of isopropyl alcohol and water. The washed product is said to have an acid number of 1.60 meq/g which also signifies inadequate removal of acidic impurities. In this case also there is no mention of the amount of chlorine in the washed PIBSA although this has to be present in significant amounts considering the use of chlorine to complete the PIBSA forming reaction. Again, EP-A-0355895 describes polyolefin substituted succinic anhydrides, eg PIBSAs, essentially free of chlorine by reaction of a polyolefin containing at least 70% of the terminal groupings in a structure having alpha olefinic bonds and a process for their production by thermal maleinisation of the polyolefin. In this document, tarry products are said to be removed from the reaction products by filtration and/or extraction with water. Notably, the acid value of the resultant products vary from 2.40 to 2.98 milli equivalent of residual acid/gram in all the Examples except Example 4 in which this value is 1.23 mmol/g. It is clear from these acid values that the PIBSA produced retained residual acids/impurities.

It has now been found that HSSAs which are substantially free of undesirable and resinous products can be produced and the corresponding succinimides produced by the reaction of these HSSAs with a polyamine perform remarkably better as detergents in gasoline and diesel fuels than the imides produced from unpurified HSSAs.

Accordingly, the present invention is a hydrocarbyl substituted succinylating agent in which the real and apparent mole ratios of succinyl groups to hydrocarbyl groups are substantially equivalent and which contains less than 250ppm of chlorine.

The mole ratios of succinyl groups to hydrocarbyl groups (which is also known as the substitution ratio or the succinylation ratio) can be calculated by measuring the base equivalent (mg KOH/g) of the succinylated product. For the purposes of the present invention, this mole ratio of succinyl groups to the hydrocarbyl groups bound to the succinyl groups is calculated from the equation below:

$$\frac{Mn \times \text{base equivalent (mg KOH/g)}}{112200 - (98 \times \text{base equivalent})} \times \frac{100}{\text{conversion}} \quad (1)$$

where the base equivalent is that of the HSSA and the conversion is the percentage conversion of the olefinic hydrocarbon to the corresponding HSSA. If the ratio described in equation (1) above is calculated using the base equivalent of the HSSA, an "apparent" substitution ratio is obtained which does not normally correspond to the "real" mole ratio of succinyl moieties to hydrocarbyl groups bound to the succinyl groups since the HSSA typically contains acidic impurities. The "real" succinylation ratio is calculated using the above equation (1) but substituting the base equivalent of the HSSA after purification eg by washing according to the following procedure.

The HSSA is diluted eg with heptane (HSSA/heptane ca. 30/70 v/v) and this solution is placed in a large separating funnel together with an aqueous solvent, eg an acetone-water mixture (ca. 30/50 v/v), the ratio of the heptane solution

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to the acetone-water mixture being about 2:1 v/v. The contents of the separating funnel are then shaken thoroughly and then allowed to separate. Two layers are formed of which the lower, aqueous layer is run off and kept. The upper, heptane layer is rewashed with a fresh aliquot of acetone-water mixture and again separated as previously. Thereafter the heptane layer is washed twice more with pure water and then separated. n-Butanol can advantageously be added to break any emulsions formed during the separation. The combined aqueous-acetone layers can be then evaporated to a volume of about 100 ml and then extracted several times with a solvent, eg petroleum ether. The ether-washed aqueous layer can be then finally separated and evaporated to dryness and heated to 170°C. The ether fractions can also be evaporated under vacuum until all the ether solvent has been removed. The residual viscous material can be combined with the original heptane layer and the solvents from this combined ether-heptane fraction removed by evaporation. The residue left behind after this step is heated eg to 180°C under vacuum. The residual solids from the evaporation of the aqueous layers are analysed to ensure that no hydrocarbyl compounds are present. The base equivalent (mg KOH/g) of the residual solids from the organic ether-heptane layers is then measured and the "real" succinylation ratio is calculated.

A feature of the HSSAs of the present invention is that these apparent and real mole ratios are substantially equivalent. By the expression "substantially equivalent" is meant here and throughout the specification that the discrepancy between the real and apparent mole ratios of succinyl groups to hydrocarbyl groups in the product is less than 0.1 and that the product is substantially free of resins and other undesirable by-products.

The novel HSSA of the present invention is typically a poly(iso)butenyl substituted succinylating agent and is suitably prepared by reacting a poly(iso) butene (hereafter "PIB"), which may be a polymer of n-butene, iso-butene or a copolymer of the two from a mixed feedstock comprising both n- and iso-butenes, with maleic anhydride (hereafter "MA"). This reaction gives rise to a poly(iso)butenyl succinic anhydride (hereafter "PIBSA"). PIBs when used for producing these PIBSAs may be conventional PIBs such as HYVIS® or PIBs which have a high terminal unsaturation, ie PIBs chains which terminate in a =CH<sub>2</sub> function. Conventional PIBs usually have less than 50% of the chains with terminal unsaturation and most commercial products have less than 20% of chains with terminal unsaturation. The preferred PIBs used for producing the PIBSAs of the present invention have >50% of the chains with terminal unsaturation. The number average molecular weight (hereafter "Mn") of the PIB when used is at least 500, and is preferably >750. A typical example of such a PIB is ULTRAVIS® (ex BP Chemicals Ltd) and a process for producing this polymer is claimed and described in our published EP-A-145235. ULTRAVIS® can be produced in grades which have a terminal unsaturation (ie =CH<sub>2</sub> end group) content of over 60% and in Mn ranging from 750-5000. Such PIBs can be produced substantially free of chlorine by the process described in our published EP-A-145235.

The PIB is reacted with MA to form the PIBSA as stated above. No chlorination of the PIB is needed to facilitate the reaction with MA. The reaction is suitably carried out in the presence of a high boiling solvent although a solvent is not essential for the PIB can be reacted with molten MA. For this reaction, it is preferable that the MA and PIB are substantially dry, ie free of water and other impurities, in order to minimise the formation of undesirable by-products.

This reaction is normally carried out in the absence of any catalyst. However, if necessary, a catalyst can be used. Examples of such catalysts include the Lewis acids eg aluminiumtrichloride or alkyl aluminium halides.

The reaction temperature for this maleinisation step is suitably from 100 to 240°C, preferably from 180 to 240°C.

The product from this maleinisation step is a mixture containing the PIBSA, maleic anhydride resins and various side products depending upon the nature of the starting PIB and whether or not the PIB is chlorinated before reaction with MA.

The HSSA of the present invention can be produced by a number of processes. A mild thermal maleinisation can give the product directly or a conventional thermal maleinisation can be followed by a purification procedure to remove any resins formed during the maleinisation. Such procedures include filtration, washing or solvent precipitation.

A preferred process involves washing with a polar solvent. It is obvious and conventional to clean a succinimide eg by a polar solvent washing in order to render the same free of unwanted impurities before being employed as a detergent or as a dispersant in eg lubricating oil or fuel compositions. However, due to the inherent dispersancy properties of the succinimide, the succinimide has a tendency to form very stable emulsions with polar solvents, especially water, thereby making the cleaning step extremely difficult. The inventive feature is to avoid trying to clean the derivative of HSSA such as eg the succinimide, but cleaning instead the HSSA used for producing the derivative, such as eg the succinimide prior to the imination reaction. In this way the polar maleic anhydride resins are removed before they are aminated. This step has the added advantage of increased efficiency of removing the maleic anhydride resins before they are aminated in that the maleic anhydride resins are more polar than the corresponding aminated maleic anhydride resin.

The cleaning procedure will be described below using crude PIBSA as a specific example of the impure HSSA. Thus, the crude PIBSA is diluted with a non-polar solvent such as eg an aromatic solvent sold commercially as HAN 8572 (ex Exxon Chemicals) and then cleaned to render it substantially free of resins and other impurities before progressing to the next stage. The effect of dilution is to enable the PIBSA to be entrained along with the hydrocarbon solvent and to facilitate filtering. In this optional step, the diluted crude PIBSA is passed through a filter bed, eg of

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diatomaceous earth filter cake, where a substantial portion of the resin and other impurities are retained and the bulk of the PIBSA is recovered as the filtrate. The filtrate so derived can be further cleaned by intimate mixing with a solvent which is preferably polar and is substantially immiscible with the hydrocarbon solvent and is capable of removing any remaining resins and other undesirable impurities in the PIBSA. Examples of such polar solvents include oxygenated solvents and water. This second cleaning step can be carried out by treating the crude PIBSA - either as such or after dilution with a hydrocarbon solvent - with a polar solvent. Where a polar solvent is used, a polar solvent phase containing substantially all of the resins and other undesirable materials can be separated from an organic hydrocarbon phase. The organic phase separated contains the cleaned PIBSA and the solvent mixture and is substantially free of any of the undesirable products or resins. The cleaning procedure can be repeated several times in order to ensure that substantially all of the impurities and resins have been removed. The end point in such a cleaning process can be detected by monitoring the acidity of the polar solvent phase until the acidity thereof is substantially constant. Such testing ensures that any acidity values in the purified PIBSA is due to the succinyl groups in the PIBSA and not due to any impurities such as unreacted MA or MA resins or other by-products associated with the PIBSA. The cleaned PIBSA can be recovered from the solvent, if desired, by removal of the hydrocarbon solvent eg by stripping or evaporation. If the PIBSA is to be further reacted with an amine to produce an imide, the organic phase from the cleaning stages can be used as such without removal of the solvent.

Thus, according to a further embodiment, the present invention is a process for the preparation of purified HSSA in which the real and apparent mole ratios of succinyl groups to hydrocarbyl groups are substantially equivalent and which contains less than 250ppm of chlorine, said process comprising:

- a. reacting a poly(iso)butene with maleic anhydride to form an HSSA and
- b. cleaning the HSSA so formed by solvent extraction to obtain the desired product.

When the HSSA is a PIBSA and when so produced is of the desired purity, ie the real and apparent mole ratios of succinyl groups to hydrocarbyl groups therein are substantially equivalent, it can be reacted with:

- a. an amine to form the corresponding succinimide,
- b. an alcohol to form the corresponding ester, or,
- c. with a metal compound such as eg the hydroxide or oxide to form the corresponding salt.

These succinimides, esters and/or salts are then used as additives for fuels or lubricants.

When the PIBSA is reacted especially with a polyamine to produce the corresponding poly(iso)butenyl succinimide, this may be a mono- or bis-succinimide and will hereafter be referred to as "PIBSI". Such PIBSIs are useful as a detergent additive in fuel and lubricating oil formulations.

This reaction between the cleaned PIBSA and the polyamine can be carried out optionally in the presence of a solvent.

The reactant polyamine used for this step is suitably an amine which has at least two basic nitrogen atoms of which at least one is a primary amino group. A polyamine substituted with -OH groups or alkoxy groups or polyoxyalkylene groups can also be used in this step. Specific examples of the polyamines that may be used include ethylene diamine, dimethylamino propylamine, triethylene tetramine, tetraethylene pentamine, N-hydroxyethyl ethylenediamine and polyether amines.

The reaction between the PIBSA and the polyamine is suitably carried out at a temperature in the range from 100 to 200°C, preferably from 50 to 190°C and a pressure from 1 to 5 bar gauge.

The relative molar ratios of the PIBSA to the polyamine in the reaction mixture is suitably in the range from 2:1 to 1:1. Irrespective of the relative ratios of the two components in this reaction, it must be ensured that the resultant product is substantially free of any unreacted polyamine before being incorporated in a fuel formulation.

According to yet another embodiment, the present invention is a derivative of an HSSA in which the hydrocarbyl substituent groups is a poly(iso)butenyl group, which derivative is an imide, an ester or a salt obtainable by a process described above and in which the real and apparent mole ratios of the succinyl groups to the poly(iso)butenyl groups is substantially equivalent and contains less than 250 ppm of chlorine.

According to a further embodiment, the present invention is a process for producing a derivative of cleaned HSSA in which the real and apparent mole ratios of the succinyl groups to poly(iso)butenyl groups are substantially equivalent and contains less than 250 ppm of chlorine, said process comprising:

- a. reacting a poly(iso)butene with maleic anhydride to form an HSSA,
- b. cleaning the HSSA so formed by solvent extraction to obtain a poly(iso)butenyl succinylating agent in which the real and apparent mole ratios of succinyl groups to poly(iso)butenyl groups is substantially the same and contains less than 250 ppm of chlorine, and

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c. reacting the cleaned succinylating agent with a reagent selected from the group consisting of:

- i. a polyamine which has at least two basic nitrogen atoms of which at least one is primary, and optionally carries hydroxy, alkoxy or polyoxyalkylene substituent groups,
- ii. an aliphatic hydroxy compound containing at least one -OH group, and
- iii. a metal hydroxide or a metal oxide,

so as to respectively form the corresponding succinimide, succinate ester or succinate salt.

The PIBSI thus produced can be used for blending into lubricating oils, or as gasoline additives optionally in a solvent and/or a carrier which may be a synthetic or a mineral oil. If the polyamine is substituted by alkoxy or polyoxyalkylene groups it may be unnecessary to use a carrier. A particular feature of the PIBSIs produced by the process of the present invention is that they are substantially free of chlorine. This is a particular advantage when such PIBSIs are used as additives for the fuels or lubricants used in internal combustion engines.

Thus, according to a yet further embodiment, the present invention is a lubricating oil or fuel composition comprising a poly(iso)butenyl succinimide in which the real and apparent mole ratios of succinyl groups to poly(iso)butenyl groups are substantially equivalent.

The present invention is further illustrated with reference to the following Examples:

**EXAMPLE 1:****1.1. Preparation of PIBSA:**

A PIB (Mn ca. 950, ULTRAVIS®10, terminal unsaturation approx. 67%, ex BP Chemicals Ltd) was reacted with a 1.99 molar excess of molten MA as follows:

The PIB was heated to 175°C and pressurised to 15 psig with nitrogen in a reactor. Molten MA was then added to the PIB over a period of about 1 hour. The reactor was then heated to 225°C during the addition whilst maintaining the reactor pressure for about 2 hours. A crude PIBSA product was formed which was stripped under vacuum for 4 hours to substantially remove excess unreacted MA. The resultant product free of unreacted MA was diluted 80/20 v/v with an aromatic hydrocarbon solvent (HAN 8572, ex Exxon Chemicals) and filtered through a bed of diatomaceous earth to remove insoluble poly-maleic anhydride resins. The resultant filtrate containing PIBSA can be used as such or after the removal of the HAN 8572 solvent for further processing. This is an example of a PIBSA which is oil-free and the final PIBSI formulation produced therefrom will also be oil-free.

The same general procedure was used to produce HYVIS® 10 PIBSA except that the reaction time for HYVIS® 10 PIBSA was four hours as against 2 hours for ULTRAVIS® 10 PIBSA.

In the case where the final PIBSI formulation is an oil-containing formulation, the dilution of the crude PIBSA product, after removal of unreacted MA, is carried out using a mineral oil which is solvent neutral grade 130/150 and this oil remains in the final PIBSI formulation substantially unchanged as does the HAN 8572 when used.

**1.2 Preparation of Purified PIBSAs:**

Oil-free ULTRAVIS®10 PIBSA (200g) as produced in Example 1.1 after filtration through a diatomaceous earth filter cake and still containing substantially all of the HAN 8572 solvent was further diluted with 1 : 1 v/v with a mixture of xylene/butanol (4:6 v/v). Water (400g) was then added to the above mixture and the mixture was stirred vigorously for 30 minutes. The mixture was transferred to a separating funnel and then allowed to stand to enable separation of aqueous and organic phases to occur. The lower aqueous layer was separated and run off. The washing procedure was repeated using further quantities of water (200g) until the acidity of the aqueous layer was unaffected by the wash. The organic layer was then distilled under vacuum to a temperature of about 180°C to remove the organic solvents and any residual water. The resultant purified PIBSA was used in subsequent preparations of the corresponding PIBSI.

**Example 2 - Preparation of Purified ULTRAVIS® 10 PIBSI (Mono-imide):**

Purified ULTRAVIS®10 PIBSA (300 g) prepared according to Example 1.2 above was charged to a round-bottomed flask rigged for atmospheric to vacuum distillation and fitted with a stirrer and dropping funnel. The flask was heated to 175°C and tetraethylene pentamine ("TEPA", 36.0 g) added via the dropping funnel over 15-20 minutes. The flask temperature was held at 175°C for a further 3 hours and vacuum applied to aid removal of water as it was formed. The product was then allowed to cool and analysed for nitrogen content. The final product had a total nitrogen content of 3.0 % w/w, a basic nitrogen content of 1.45 % w/w and an alkalinity value of 58.3 mg KOH/g.

**EP 0 602 863 B1**Example 3 - Preparation of Purified ULTRAVIS® 10 PIBSI (Bis succinimide):

Purified ULTRAVIS® 10 PIBSA (300 g) produced according to the process of Example 1.2 was charged to a 1 litre round-bottomed flask rigged for distillation and fitted with a stirrer and dropping funnel. The flask contents were heated to 175°C and triethylene tetramine ("TETA", 16 g) added dropwise over 15-20 minutes. The flask contents were maintained at 175°C under partial vacuum for a further 3 hours. Water formed during the reaction was distilled off under vacuum. The resultant PIBSI product had a total nitrogen content of 1.88% w/w and an alkalinity value of 23.8 mg KOH/g.

In a series of Comparative Tests ( "CT", not according to the invention), further samples of PIBSI were prepared which had not been water-washed to remove any residual resinous or other undesirable impurities. These are described below:

CT1. Preparation of unclean ULTRAVIS®10 PIBSI (Mono succinimide):

ULTRAVIS®10 PIBSA (450g, PIBSA No. 76 mg KOH/g) prepared as in Example 1.1 above, diluted 80/20 v/v with an aromatic hydrocarbon solvent (HAN 8572, ex Exxon Chemicals) and filtered through a diatomaceous earth filter cake was charged to a 1 litre flask fitted with a stirrer, dropping funnel, Dean and Stark receiver, condenser and nitrogen purge. Triethylene tetramine ("TETA", 22.25g) was charged to the dropping funnel. The TETA was then added dropwise over 15-20 minutes. The flask temperature was allowed to rise slowly to 175°C over 3 hours and the liberated water collected in the Dean and Stark trap. This gave a PIBSI with a basic nitrogen content of 0.73% w/w, a total nitrogen content of 1.7% w/w and an alkalinity value of 29.5 mg KOH/g.

In addition to the above, a sample of conventional PIBSI made from HYVIS® 10 polybutene and commercially sold as ADX 201 (mono-imide) by Adibis Ltd UK was also used in the Comparative Tests.

Example 4 - Engine Test Data:

Samples of the PIBSI prepared according to the Examples and Comparative Tests above were tested to evaluate their performance by replacing on a weight for weight basis the detergent component of a modern high-performance engine inlet system detergent package. The detergent package used was ADX3866, a commercial package available from BP Chemicals (Additives) Ltd.

The results are given in the Tables below:

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**TABLE 1**

Test No	Additive Used	Package Treatment level (ppm w/w)	Engine Test	Valve Deposit mg/valve)
A	Base Fuel (8A85)	0	Opel Kadett	397
B	Bis-imide from CT1	500	Opel Kadett	172
C	Bis-imide from Ex 3	500	Opel Kadett	75
D	Bisimide C water-washed	500	Opel Kadett	61

**TABLE 2**

Test No	Additive Used	Package Treatment level (ppm w/w)	Engine Test	Valve Deposit mg/valve)
E	Base Fuel (83A87)	0	M102E	296
F	ADX201	1000	M102E	57
G	Product of Example 2	1000	M102E	10

The results in Tables 1 and 2 show that the succinimides of the present invention are significantly superior to those of prior art and that purifying the final imide again, eg by washing, does not significantly further improve performance.

Example 5 - Further preparations of cleaned HSSAs derived from HYVIS® and ULTRAVIS®:

Samples of HYVIS® polybutene (200g) and ULTRAVIS® polybutene (200g) were separately subjected to a malein-



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isation reaction using in each case a molar excess of maleic anhydride using reaction conditions shown in Table 3 below. The crude products (PIBSAs) in each case were stripped of excess unreacted maleic anhydride and the stripped product in each case dissolved in heptane (800 ml). Celite filter aid was mixed with each of the heptane solutions of the PIBSAs which were then stirred for several minutes. Each of the mixtures were then separately filtered through a  
5 Celite filter cake. The resultant filtrates containing the respective PIBSA in heptane were washed twice with a water/acetone mixture (60/40 v/v, 300 ml) and after each washing the mixed solutions were allowed to separate into an organic phase and an aqueous phase. In each case, the aqueous layer was discarded and the organic phase recovered after the second washing was stripped of the solvent. The resultant solvent-free product was heated in each case to 180°C in a rotary evaporator under a strong vacuum for one hour to form a cleaned PIBSA of the present invention.  
10 The procedure was repeated to produce a further aliquot each of the cleaned PIBSAs from HYVIS® and ULTRAVIS® so as to obtain a combined weight of PIBSAs in each case of about 500g. The analysis of the product obtained is shown below in Table 3:

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TABLE 3

PIB	Mn	MA added mol/mol PIB	Reaction Cond Hr °C	PIBSA Conv. %	PIBSA* Acidity mg KOH/g	Succinylation Ratio of purified PIBSA**
HYVIS®	923	3	4 235	70.41	78.23	0.98
ULTRAVIS	994	3	4 235	88.7	90.0	0.98

\*PN (PIBSA No.)  
\*\* This was found to be unchanged by further washing

**EP 0 602 863 B1****Example 6 - Preparation of Succinimides:**

The washed HYVIS® PIBSA (285.5g) from Example 5 above was reacted with TETA (14.83g) for 3 hours at 175-180°C in a stirred reactor provided with a nitrogen sparge. The by-product water from the reaction mixture was removed by the nitrogen sparge.

Similarly, the washed ULTRAVIS® PIBSA from Example 5 was reacted with TETA for 3 hours at 175-180°C and the by-product was removed, as previously, with a nitrogen sparge.

The products from both these reactions were bis-succinimides which were used as detergents in a commercial formulation ADX3866 (ex Adibis Ltd) used for engine testing. The treatment levels used and results achieved are tabulated in Table 4 below.

**CT2 - Preparation of succinimides (not according to the invention):**

A further set of PIBSAs were made from HYVIS® and ULTRAVIS® following the process described in Example 5 above but the products from the initial reaction of polybutene with maleic anhydride were not diluted in heptane and were not washed. Each product was separately stripped of excess maleic anhydride and filtered at 100°C through a pre-prepared Celite filter cake. The acidities (PIBSA No. mg KOH/g) of the filtered PIBSAs were measured in each case and they were then reacted separately with an appropriate quantity of TETA [Wt of TETA added = (wt of PIBSA x PIBSA No. x 146 x 0.5)/112200] to make the corresponding bis-succinimide. The reaction in each case was carried out as previously at 175-180°C for 3 hours, and by-product water was removed by a nitrogen sparge. The resultant bis-succinimides were used as detergent in a commercial formulation ADX3866 (ex Adibis Ltd) used for engine testing as previously. The treatment levels used and the results achieved are tabulated in Table 4 below:

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**TABLE 4**

Detergent in Formulation	Package Treatment level (ppm)	Engine tested	Valve Deposit (mg/valve)	Valve Rating	Manifold Rating
Unwashed HYVIS® bis-succinimide	500	Opel Kadett	106	8.99	9.44
Washed HYVIS® bis-succinimide	500	Opel Kadett	49	8.8	9.94
Unwashed ULTRAVIS® bis-succinimide	500	Opel Kadett	62	9.34	8.74
Washed ULTRAVIS® bis-succinimide	500	Opel Kadett	35	9.31	9.69

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The above results show that formulations which use succinimides from purified PIBSAs of the present invention perform significantly better than those of the prior art in respect of valve cleanliness.

**EXAMPLE 7:**

A PIBSA in which the real and apparent mole ratios of the poly(iso)butenyl groups to succinyl groups were substantially equivalent was prepared by reacting HYVIS® polybutene (200g, Mn 2045) with maleic anhydride (28 g) at 235°C for six hours. The apparent mole ratio of the reaction product after stripping excess maleic anhydride and filtration in mineral oil through a Celite filter bed in mineral oil was 0.95. The real mole ratio after a washing procedure as described in the text above was found to be 0.89.

Thus, the products before washing had substantially the same mole ratio as that of the washed product. This shows that the products of the present invention can be produced by processes which do not involve washing.

**Claims**

1. A hydrocarbyl substituted succinylating agent (HSSA) in which the discrepancy between the real and apparent mole ratios of the succinyl groups to hydrocarbyl groups is less than 0.1 and which contains less than 250 ppm of chlorine.

2. A process for producing an HSSA in which the discrepancy between the real and apparent mole ratios of the succinyl groups to hydrocarbyl groups is less than 0.1 and which contains less than 250 ppm of chlorine, said process comprising:

a. reacting a poly(iso)butene with maleic anhydride to form an HSSA, and  
b. cleaning the HSSA so formed by solvent extraction to obtain the desired product, said solvent extraction step comprising:

i. optionally diluting the HSSA with a non-polar solvent,

ii. filtering the diluted HSSA from (i) to remove the bulk of the resin and other impurities as a residue and a solution of the HSSA as a filtrate substantially free of resins and other impurities,

iii. intimately mixing the filtrate from (ii) or, when optional step (i) is not used, the undiluted HSSA, with a polar solvent capable of removing any remaining resins and other impurities in the filtrate or the HSSA and is substantially immiscible with the non-polar solvent in the filtrate, or, the HSSA,

iv. separating the polar solvent phase containing the resin and other impurities from the non-polar diluent phase containing the HSSA, or, when step (i) is not used, the HSSA, and

v. recovering the cleaned HSSA therefrom as such, or, when step (i) is used, by removing the non-polar diluent phase containing the same.

3. A process according to Claim 2 wherein the poly(iso)butene is a polymer of n-butene, isobutene or a copolymer of the two from a mixed feedstock comprising both n- and iso-butenes.

4. A process according to Claim 2 or 3 wherein the poly(iso)butene has >50% of the chains terminally unsaturated.

5. A process according to any one of the preceding Claims 2-4 wherein the poly(iso)butene has a number average molecular weight of at least 500.

6. A process according to any one of the preceding Claims 2-5 wherein step (a) is carried out by reacting poly(iso)butene with maleic anhydride in amounts ranging from 1:0.5 to 1:5 molar ratio of poly(iso)butene to maleic anhydride.

7. A process according to any one of the preceding Claims 2-6 wherein step (a) is carried out in the absence of a catalyst.

8. A process according to any one of the preceding Claims 2-7 wherein step (a) is carried out at a temperature from 100-240°C.

9. A process according to any one of the preceding Claims 2-8 wherein the cleaning step (b) is carried out using a

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solvent comprising a polar solvent.

10. A process according to Claim 9 wherein the polar solvent is selected from oxygenated solvents and water.

5 11. A process according to Claim 9 or 10 wherein the cleaning step (b) is carried out by partitioning the reaction products from step (a) in a mixture of solvents comprising a hydrocarbon solvent and a polar solvent whereby the mixture of solvents separates into a polar solvent phase and an organic hydrocarbon phase such that the polar solvent phase contains substantially all of the resins and other undesirable materials whereas the organic hydrocarbon solvent phase contains the cleaned succinate substantially free of any undesirable resins or undesirable by-products.

10 12. A process for producing a derivative of cleaned HSSA in which the discrepancy between the real and apparent mole ratios of the succinyl groups to hydrocarbyl groups is less than 0.1 and contains less than 250 ppm of chlorine, said process comprising:

15 a. reacting a poly(iso)butene with maleic anhydride to form an HSSA according to any one of the preceding Claims 2-8,

b. cleaning the HSSA so formed by solvent extraction according to any one of the preceding Claims 9-11 to obtain a poly(iso)butenyl succinylating agent in which the real to apparent mole ratios of succinyl groups to poly(iso)butenyl groups is substantially the same and contains less than 250 ppm of chlorine, and

20 c. reacting the cleaned succinylating agent with a reagent selected from the group consisting of:

i. a polyamine which has at least two basic nitrogen atoms of which at least one is primary, and optionally carries hydroxy, alkoxy or polyoxyalkylene substituent groups,

25 ii. an aliphatic hydroxy compound containing at least one -OH group, and

iii. a metal hydroxide or a metal oxide,

so as to respectively form the corresponding succinimide, succinate ester or succinate salt.

30 13. A process according to Claim 12 wherein the polyamine used is selected from the group consisting of ethylene diamine, dimethylamino propylamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine and N-hydroxyethyl ethylenediamine.

35 14. A process according Claim 12 or 13 wherein the mole ratio of the cleaned succinylating agent to the polyamine for step (c) is in the range from 2:1 to 1:1.

15. A process according to any one of the preceding Claims 12-14 wherein the reaction temperature for step (c) is from 100 to 200°C and the reaction pressure is from 1 to 5 bar gauge.

40 16. A derivative of an HSSA, said HSSA being according to claim 1 and the hydrocarbyl substituent group being a poly(iso)butenyl group, which derivative is an imide, an ester or a salt obtainable by a process according to any one of the preceding Claims 12-15 and in which the discrepancy between the real and apparent mole ratios of the succinyl groups to hydrocarbyl groups is less than 0.1 and contains less than 250 ppm of chlorine.

45 17. A derivative according to Claim 16 wherein the poly(iso)butenyl group in each derivative is derived from a poly(iso)butene which has at least 50% of the chains with terminal unsaturation.

50 18. A lubricating oil or fuel composition comprising a derivative of an HSSA in which the hydrocarbyl group is a poly(iso)butenyl group and the discrepancy between the real and apparent mole ratios of the succinyl groups to hydrocarbyl groups is less than 0.1 and contains less than 250 ppm of chlorine according to Claim 16 or 17, said HSSA being prepared by a process according to any one of the preceding Claims 2-11 and said derivative being selected from the group consisting of an imide, an ester, and a salt of HSSA prepared by a process according to any one of the preceding Claims 12-15.

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#### Patentsprüche

1. Kohlenwasserstoff-substituiertes Succinylierungsmittel (HSSA), bei dem die Diskrepanz zwischen dem wirklichen

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und dem scheinbaren Molverhältnis der Succinylgruppen zu den Kohlenwasserstoffgruppen weniger als 0,1 beträgt und das weniger als 250 ppm Chlor enthält.

- 5 2. Verfahren zur Herstellung eines HSSA, bei dem die Diskrepanz zwischen dem wirklichen und dem scheinbaren Molverhältnis der Succinylgruppen zu den Kohlenwasserstoffgruppen weniger als 0,1 beträgt und das weniger als 250 ppm Chlor enthält, wobei das Verfahren umfaßt:
- 10 a. Umsetzen eines Poly(iso)butens mit Maleinsäureanhydrid zu einem HSSA und  
b. Reinigen des so gebildeten HSSA durch Lösungsmittelextraktion zu dem gewünschten Produkt, wobei der Lösungsmittel-Extraktionsschritt umfaßt:
- 15 i. gegebenenfalls Verdünnen des HSSA mit einem unpolaren Lösungsmittel,  
ii. Filtrieren des verdünnten HSSA von (i) zur Entfernung des Hauptteils des Harzes und weiterer Verunreinigungen als Rückstand und eine Lösung des HSSA als im wesentlichen von Harz und weiteren Verunreinigungen freies Filtrat,  
15 iii. inniges Vermischen des Filtrats von (ii) oder, wenn der mögliche Schritt (i) nicht verwendet wird, des unverdünnten HSSA, mit einem polaren Lösungsmittel, das verbliebene Harze und weitere Verunreinigungen in dem Filtrat oder dem HSSA entfernen kann und im wesentlichen mit unpolarem Lösungsmittel in dem Filtrat, oder dem HSSA, nicht mischbar ist,  
20 iv. Trennen der polaren Lösungsmittelphase, die das Harz und weitere Verunreinigungen enthält, von der unpolaren verdünnten Phase, die das HSSA enthält, oder, wenn Schritt (i) nicht eingesetzt wird, dem HSSA, und  
v. Gewinnen des gereinigten HSSA daraus als solches oder, wenn Schritt (i) verwendet wird, durch Entfernen der unpolaren verdünnten Phase, die dasselbe enthält.
- 25 3. Verfahren nach Anspruch 2, wobei das Poly(iso)buten ein Polymer von n-Buten, Isobuten oder einem Copolymer der zwei aus einer Mischbeschickung, die sowohl n- als auch Isobuten umfaßt, ist.
- 30 4. Verfahren nach Anspruch 2 oder 3, wobei das Poly(iso)buten >50% der Ketten endständig ungesättigt aufweist.
5. Verfahren nach einem der vorangehenden Ansprüche 2-4, wobei das Poly(iso)buten ein zahlenmittleres Molekulargewicht von mindestens 500 aufweist.
- 35 6. Verfahren nach einem der vorangehenden Ansprüche 2-5, wobei Schritt (a) durch Umsetzen von Poly(iso)buten mit Maleinsäureanhydrid in Mengen in einem Molverhältnis von Poly(iso)buten zu Maleinsäureanhydrid im Bereich 1:0,5 bis 1:5 ausgeführt wird.
7. Verfahren nach einem der vorangehenden Ansprüche 2-6, wobei Schritt (a) in Abwesenheit eines Katalysators ausgeführt wird.
- 40 8. Verfahren nach einem der vorangehenden Ansprüche 2-7, wobei Schritt (a) bei einer Temperatur von 100-240°C ausgeführt wird.
- 45 9. Verfahren nach einem der vorangehenden Ansprüche 2-8, wobei der Reinigungsschritt (b) unter Verwendung eines Lösungsmittels, umfassend ein polares Lösungsmittel, ausgeführt wird.
10. Verfahren nach Anspruch 9, wobei das polare Lösungsmittel ausgewählt ist aus sauerstoffhaltenden Lösungsmitteln und Wasser.
- 50 11. Verfahren nach Anspruch 9 oder 10, wobei der Reinigungsschritt (b) durch Verteilen der Reaktionsprodukte von Schritt (a) in einem Gemisch von Lösungsmitteln, umfassend ein Kohlenwasserstoff-Lösungsmittel und ein polares Lösungsmittel, ausgeführt wird, wodurch das Gemisch von Lösungsmitteln in eine polare Lösungsmittelphase und eine organische Kohlenwasserstoffphase getrennt wird, so daß die polare Lösungsmittelphase im wesentlichen die gesamten Harze und weitere unerwünschte Materialien enthält, wohingegen die organische Kohlenwasserstofflösungsmittelphase das gereinigte Succinat, das im wesentlichen frei von unerwünschten Harzen oder unerwünschten Nebenprodukten ist, enthält.
- 55 12. Verfahren zur Herstellung eines Derivats von gereinigtem HSSA, wobei die Diskrepanz zwischen dem wirklichen

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und scheinbaren Molverhältnis der Succinylgruppen zu den Kohlenwasserstoffgruppen weniger als 0,1 beträgt und das weniger als 250 ppm Chlor enthält, wobei das Verfahren umfaßt:

- 5 a. Umsetzen eines Poly(iso)butens mit Maleinsäureanhydrid zu einem HSSA nach einem der vorangehenden Ansprüche 2-8,
- b. Reinigen des so gebildeten HSSA durch Lösungsmittelextraktion nach einem der vorangehenden Ansprüche 9-11 zu einem Poly(iso)butenyl-Succinylierungsmittel, bei dem das wirkliche und scheinbare Molverhältnis von Succinylgruppen zu den Poly(iso)butenylgruppen im wesentlichen dasselbe ist und das weniger als 250 ppm Chlor enthält, und
- 10 c. Umsetzen des gereinigten Succinylierungsmittels mit einem Reagenz, ausgewählt aus der Gruppe, bestehend aus:
  - i. einem Polyamin, das mindestens zwei basische Stickstoffatome enthält, wobei mindestens eines davon primär ist, und gegebenenfalls Hydroxy-, Alkoxy- oder Polyoxyalkylensubstituentengruppen trägt,
  - 15 ii. einer aliphatischen Hydroxyverbindung, die mindestens eine Gruppe -OH enthält, und
  - iii. einem Metallhydroxid oder einem Metalloxid,

so daß sich Succinimid, Bernsteinsäureester bzw. Succinatsalz bildet.

- 20 13. Verfahren nach Anspruch 12, wobei das verwendete Polyamin ausgewählt ist aus der Gruppe, bestehend aus Ethylendiamin, Dimethylaminopropylamin, Diethylentriamin, Triethylentetramin, Tetraethylenpentamin und N-Hydroxyethylethylendiamin.
14. Verfahren nach Anspruch 12 oder 13, wobei das Molverhältnis des gereinigten Succinylierungsmittels zu dem Polyamin für Schritt (c) im Bereich 2:1 bis 1:1 liegt.
- 25 15. Verfahren nach einem der vorangehenden Ansprüche 12-14, wobei die Reaktionstemperatur für Schritt (c) 100 bis 200°C beträgt und der Reaktionsdruck 1 bis 5 Bar Überdruck ist.
- 30 16. Derivat eines HSSA, wobei das HSSA nach Anspruch 1 definiert ist und die Kohlenwasserstoff-Substituentengruppe eine Poly(iso)butenylgruppe ist, wobei das Derivat ein Imid, einen Ester oder ein Salz darstellt, erhältlich durch ein Verfahren nach einem der vorangehenden Ansprüche 12-15, und wobei die Diskrepanz zwischen dem wirklichen und dem scheinbaren Molverhältnis der Succinylgruppen zu den Kohlenwasserstoffgruppen weniger als 0,1 beträgt und das weniger als 250 ppm Chlor enthält.
- 35 17. Derivat nach Anspruch 16, wobei die Poly(iso)butenylgruppe in jedem Derivat abgeleitet ist von einem Poly-(iso)buten, das mindestens 50% der Ketten mit endständiger Ungesättigtheit versehen aufweist.
- 40 18. Schmieröl- oder Kraftstoffzusammensetzung, umfassend ein Derivat eines HSSA, bei dem die Kohlenwasserstoffgruppe eine Poly(iso)butenylgruppe ist und die Diskrepanz zwischen dem wirklichen und dem scheinbaren Molverhältnis der Succinylgruppen zu den Kohlenwasserstoffgruppen weniger als 0,1 beträgt und das weniger als 250 ppm Chlor enthält, nach Anspruch 16 oder 17, wobei das HSSA durch ein Verfahren nach einem der vorangehenden Ansprüche 2-11 hergestellt ist und das Derivat ausgewählt ist aus der Gruppe, bestehend aus einem Imid, Ester oder Salz von HSSA, hergestellt durch ein Verfahren nach einem der vorangehenden Ansprüche 12-15.
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**Revendications**

- 50 1. Agent succinylant substitué par un hydrocarbyle (HSSA), dans lequel l'écart entre les rapports molaires réel et apparent des groupes succinyles aux groupes hydrocarbyles est inférieur à 0,1 et qui contient moins de 250 ppm de chlore.
2. Procédé de production d'un HSSA dans lequel l'écart entre les rapports molaires réel et apparent des groupes succinyles aux groupes hydrocarbyles est inférieur à 0,1 et qui contient moins de 250 ppm de chlore, ce procédé comprenant :
  - a. la réaction d'un poly(iso)butène avec de l'anhydride maléique pour la formation d'un HSSA, et
  - 55 b. la purification du HSSA ainsi formé par extraction par solvant pour l'obtention du produit désiré, cette étape



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d'extraction par solvant comprenant :

- i. facultativement, la dilution du HSSA dans un solvant non polaire,
  - 5 ii. la filtration du HSSA dilué provenant de (i) pour l'élimination du gros de la résine et des autres impuretés comme résidu et l'obtention d'une solution du HSSA en tant que filtrat sensiblement exempt de résines et d'autres impuretés,
  - iii. le mélange intime du filtrat provenant de (ii) ou, lorsque l'étape facultative (i) n'est pas utilisée, du HSSA non dilué, avec un solvant polaire capable d'éliminer les résines et autres impuretés restantes éventuelles du filtrat ou du HSSA et pratiquement non miscible avec le solvant non polaire du filtrat ou le HSSA,
  - 10 iv. la séparation de la phase solvant polaire contenant la résine et les autres impuretés de la phase diluant non polaire contenant le HSSA ou, si l'étape (i) n'est pas utilisée, du HSSA, et
  - v. la récupération du HSSA purifié de celle-ci tel quel ou, si l'étape (i) est utilisée, par élimination de la phase diluant non polaire contenant celui-ci.
- 15 3. Procédé selon la revendication 2, dans lequel le poly(iso)butène est un polymère de n-butène ou d'isobutène ou un copolymère des deux provenant d'une matière première mixte comprenant à la fois du n-butène et de l'isobutène.
- 20 4. Procédé selon l'une des revendications 2 et 3, dans lequel le poly(iso)butène a plus de 50 % de chaînes à insaturation terminale.
5. Procédé selon l'une quelconque des revendications 2 à 4 précédentes, dans lequel le poly(iso)butène a un poids moléculaire moyen en nombre d'au moins 500.
- 25 6. Procédé selon l'une quelconque des revendications 2 à 5 précédentes, dans lequel l'étape (a) est exécutée par réaction de poly(iso)butène avec de l'anhydride maléique en quantités correspondant à un rapport molaire du poly(iso)butène à l'anhydride maléique de 1/0,5 à 1/5.
- 30 7. Procédé selon l'une quelconque des revendications 2 à 6 précédentes, dans lequel l'étape (a) est exécutée en l'absence de catalyseur.
8. Procédé selon l'une quelconque des revendications 2 à 7 précédentes, dans lequel l'étape (a) est exécutée à une température de 100 à 240 °C.
- 35 9. Procédé selon l'une quelconque des revendications 2 à 8 précédentes, dans lequel l'étape de purification (b) est exécutée avec un solvant comprenant un solvant polaire.
10. Procédé selon la revendication 9, dans lequel le solvant polaire est choisi parmi les solvants oxygénés et l'eau.
- 40 11. Procédé selon l'une des revendications 9 et 10, dans lequel on exécute l'étape de purification (b) en répartissant les produits de réaction provenant de l'étape (a) dans un mélange de solvants comprenant un solvant hydrocarbure et un solvant polaire, de sorte que ce mélange de solvants se divise en une phase solvant polaire et une phase hydrocarbure organique de façon que la phase solvant polaire contienne pratiquement toutes les résines et les autres matières indésirables, tandis que la phase solvant hydrocarbure organique contient le succinate purifié
- 45 pratiquement exempt de résines indésirables ou de sous-produits indésirables.
12. Procédé de production d'un dérivé de HSSA purifié dans lequel l'écart entre les rapports molaires réel et apparent des groupes succinyles aux groupes hydrocarbyles est inférieur à 0,1 et qui contient moins de 250 ppm de chlore, ce procédé comprenant :
  - a. la réaction d'un poly(iso)butène avec de l'anhydride maléique pour la formation d'un HSSA selon l'une des revendications 2 à 8 précédentes,
  - b. la purification du HSSA ainsi formé par extraction par solvant selon l'une des revendications 9 à 11 précédentes pour l'obtention d'un agent succinylant poly(iso)butényle dans lequel les rapports molaires réel et apparent des groupes succinyles aux groupes poly(iso)butényles sont sensiblement les mêmes et qui contient
  - 50 moins de 250 ppm de chlore, et
  - c. la réaction de l'agent succinylant purifié avec un réactif choisi dans le groupe constitué :
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- i. d'une polyamine qui a au moins deux atomes d'azote de base dont au moins un est primaire, et porte facultativement des groupes substituants hydroxy, alkoxy ou polyoxyalkylènes,
- ii. d'un composé hydroxy aliphatique contenant au moins un groupe -OH, et
- iii. d'un hydroxyde métallique ou d'un oxyde métallique,

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pour la formation respectivement du succinimide, du succinate-ester ou du succinate-sel correspondant.

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13. Procédé selon la revendication 12, dans lequel la polyamine utilisée est choisie dans le groupe constitué de l'éthylènediamine, la diméthylaminopropylamine, la diéthylènetriamine, la triéthylènetétramine, la tétraéthylènepentamine et la N-hydroxyéthyléthylènediamine.

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14. Procédé selon l'une des revendications 12 et 13, dans lequel le rapport molaire de l'agent succinylant purifié à la polyamine pour l'étape (c) est compris entre 2:1 et 1:1.

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15. Procédé selon l'une des revendications 12 à 14 précédentes, dans lequel la température de réaction pour l'étape (c) est de 100 à 200 °C et la pression de réaction est de 1 à 5 bars manométriques.

16. Dérivé d'un HSSA, ce HSSA étant conforme à la revendication 1 et le groupe substituant hydrocarbyle étant un groupe poly(iso)butényle, lequel dérivé est un imide, un ester ou un sel pouvant être obtenu par un procédé selon l'une quelconque des revendications 12 à 15 précédentes, et dans lequel dérivé l'écart entre les rapports molaires réel et apparent des groupes succinyles aux groupes hydrocarbyles est inférieur à 0,1, et lequel dérivé contient moins de 250 ppm de chlore.

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17. Dérivé selon la revendication 16, dans lequel le groupe poly(iso)butényle de chaque dérivé est dérivé d'un poly(iso)butène qui a au moins 50 % de chaînes à insaturation terminale.

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18. Composition d'huile lubrifiante ou de carburant comprenant un dérivé d'un HSSA dans lequel le groupe hydrocarbyle est un groupe poly(iso)butényle et l'écart entre les rapports molaires réel et apparent des groupes succinyles aux groupes hydrocarbyles est inférieur à 0,1 et qui contient moins de 250 ppm de chlore selon l'une des revendications 16 et 17, le HSSA étant préparé par un procédé selon l'une des revendications 2 à 11 précédentes et le dérivé étant choisi dans le groupe constitué d'un imide, d'un ester et d'un sel de HSSA préparés par un procédé selon l'une quelconque des revendications 12 à 15 précédentes.

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